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Title:	Application of the time-dependent close-coupling method to ionization of multi-electron atoms
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Application of the time-dependent close-coupling method to ionization of multi-electron atoms

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Plan of talk

- Introduction
- Two-photon double ionization of the Li
- Electron-impact ionization of Na and Mg
- Summary
- Future plans

Introduction

- Time-dependent close-coupling (TDCC) method.
- Full-dimensionality numerical integration of the two/three-electron time-dependent Schrödinger equation.
- Applicable to a range of collision processes (photon, electron or ion impact).
- Accurate over a wide range of impact energies.

Two-photon double ionization of lithium

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Motivation

- Provides guidance and support for recent measurements made at FLASH (Schuricke/Dorn).

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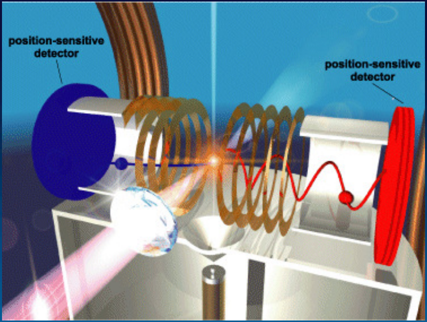
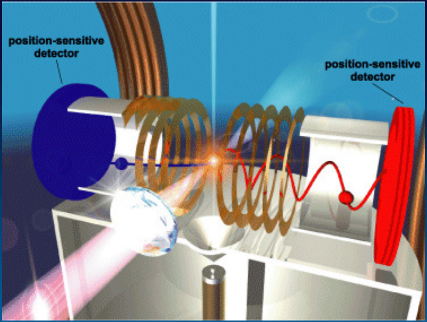
- Such comparison requires accurate resolution of angular degrees of freedom and electron momenta.
- Provide first angular-resolved calculations for two-photon double ionization of Li.
- Application of TDCC codes to treatment of more than two photon absorptions (above-threshold ionization).

FLASH experiments



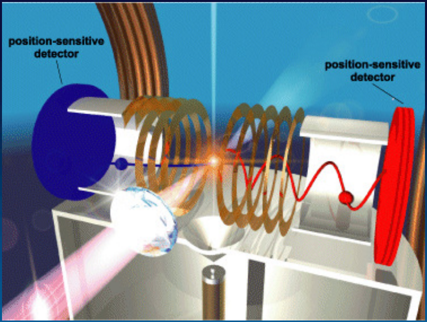
- COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy) - Separate collection of ions and electrons.

FLASH experiments



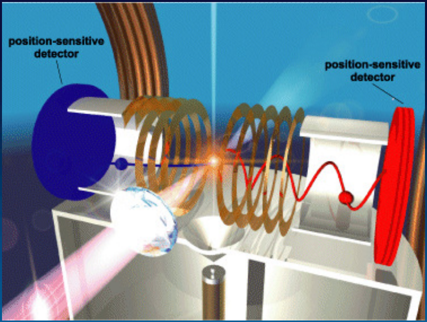
- COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy) - Separate collection of ions and electrons.
- Measured time of flight and position of ions/electrons allow momenta to be determined.

FLASH experiments



- Two-photon double ionization of Li at 50 eV (direct) and 59 eV (intermediate excitation of the 1s2s2p state).

FLASH experiments



- Two-photon double ionization of Li at 50 eV (direct) and 59 eV (intermediate excitation of the 1s2s2p state).
- Li ($1s^2 2s$) + $n\hbar\omega \rightarrow$ Li^{2+}(1s) + $2e^-$.

Theory

Time-dependent close-coupling method

- Full-dimensionality solution of the two-electron TDSE:

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r}_1,\mathbf{r}_2,t) = H\Psi(\mathbf{r}_1,\mathbf{r}_2,t).$$

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- Finite-difference/basis-set expansion for each spin state:

$$\Psi^S(\mathbf{r}_1,\mathbf{r}_2,t) = \sum_{l_1,l_2,L} \frac{P^{LS}_{l_1 l_2}(r_1,r_2,t)}{r_1 r_2}|l_1 l_2 L\rangle.$$

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- Two active electrons move in the field of a frozen 1s Li^{2+} orbital.

TDCC method

- Angular reduction of the two-electron time-dependent Schrödinger equation yields

$$i\frac{\partial}{\partial t}P^{LS}_{l_1 l_2}(r_1,r_2,t) = T_{l_1 l_2}P^{LS}_{l_1 l_2}(r_1,r_2,t) + \sum_{l_1',l_2'} V^{L}_{l_1 l_2,l_1' l_2'}P^{LS}_{l_1' l_2'}(r_1,r_2,t) + \sum_{l_1',l_2',L'} W^{L,L'}_{l_1 l_2,l_1' l_2'}P^{L'S}_{l_1' l_2'}(r_1,r_2,t).$$

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$$
\begin{aligned}
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& + \sum_{l'_1, l'_2} V^{L}_{l_1 l_2, l'_1 l'_2} P^{LS}_{l'_1 l'_2}(r_1, r_2, t) \\
& + \sum_{l'_1, l'_2, L'} W^{L,L'}_{l_1 l_2, l'_1 l'_2} P^{L'S}_{l'_1 l'_2}(r_1, r_2, t).
\end{aligned}
$$

- Initial state obtained through relaxation of the field-free time-dependent Schrödinger equation in imaginary time.

Double ionization of lithium at 50 eV

Experimental considerations

- FLASH laser pulse length $\sim$ 10 fs.
- Pulses display chaotic behaviour.
- Laser intensity will vary strongly over time.
- Intensity range: $5 \times 10^{13} \leq I \leq 5 \times 10^{15}$ (W/cm^2).

Triple-differential cross sections

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Triple-differential cross sections

- Momentum-space wavefunction, $\mathcal{P}^{LS}_{l_1 l_2}(\mathbf{k}_1, \mathbf{k}_2)$, obtained via projection of position-space wavefunction onto product of Li^{2+} continuum orbitals at the end of the laser pulse.

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- Momentum-space wavefunction, $\mathcal{P}^{LS}_{l_1 l_2}(\mathbf{k}_1, \mathbf{k}_2)$, obtained via projection of position-space wavefunction onto product of Li^{2+} continuum orbitals at the end of the laser pulse.

- Appropriate integration of momentum-space wavefunction over all momenta yields the TDCS:

$$\frac{\mathrm{d}^3\sigma}{\mathrm{d}E_1 \mathrm{d}\Omega_1 \mathrm{d}\Omega_2} =$$

$$\frac{1}{k_1 k_2}\left(\frac{\omega}{I}\right)^N \frac{1}{T_{\mathrm{eff}}} \int_0^\infty \mathrm{d}k_1 \int_0^\infty \mathrm{d}k_2 \, \delta\left[\alpha - \tan^{-1}\left(\frac{k_2}{k_1}\right)\right]$$

$$\times \sum_S w_S \left| \sum_{l_1, l_2, L} (-i)^{l_1 + l_2} e^{i(\sigma_{l_1} + \sigma_{l_2})} \mathcal{P}^{LS}_{l_1 l_2}(k_1, k_2) | l_1 l_2 L \rangle \right|^2 .$$

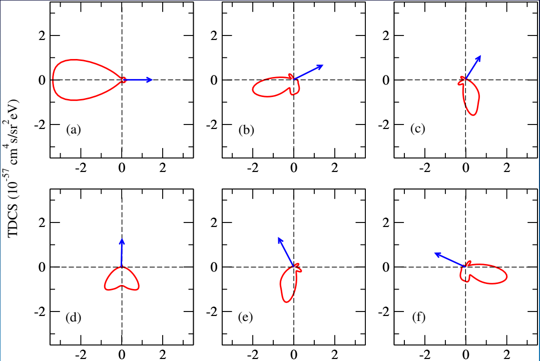
Triple-differential cross sections

- Energy of outgoing electron pair may be partitioned arbitrarily for a given total excess energy.
- Coplanar detection geometry ($\phi_1 = \phi_2 = 0°$) used to reduce angular variables from four to two.
- Polar angle of one electron is fixed, the other varied (relative to the laser polarization axis).

Triple-differential cross sections

1s2s ^{1}S initial state, equal energy sharing

$\omega = 50$ eV, $I = 5 \times 10^{14}$ Wcm^{-2}



- Anti-parallel emission also dominant for $\theta_1 = 0°, 30°$ and $\theta_1 = 150°$, with additional near-perpendicular emissions.

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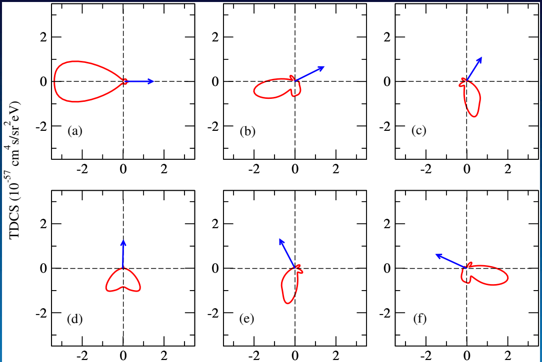

- Near-perpendicular emission dominant for $\theta_1 = 60°$ and $\theta_1 = 120°$.

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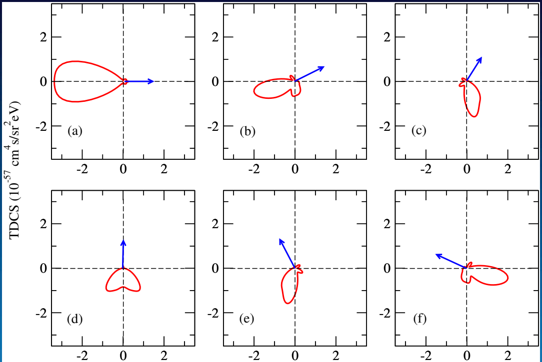

- Emission at a mutual angle $\theta_{12} = 120^\circ$ dominates for $\theta_1 = 90^\circ$.

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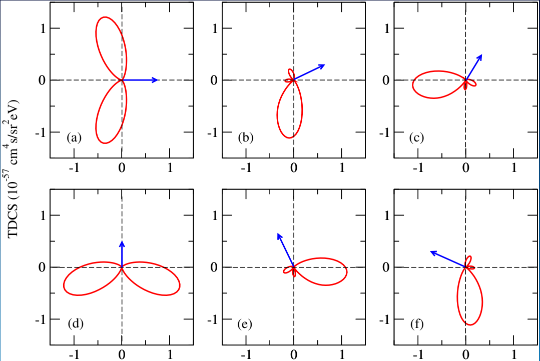

- Anti-parallel emission avoided for all values of θ_1.

Triple-differential cross sections

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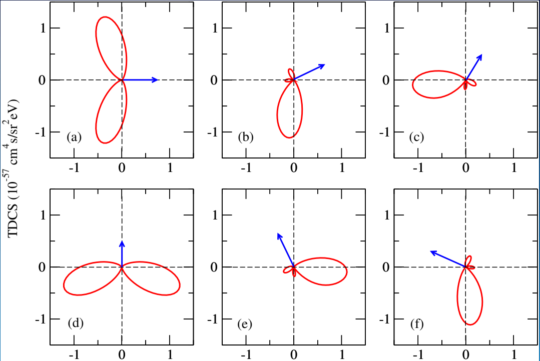
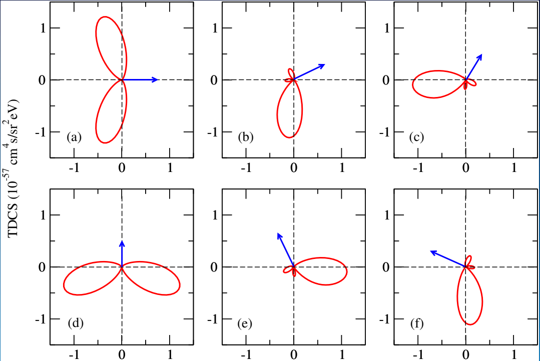


- Dominant emission at $\theta_{12} \simeq 120°$ at all angles.

Triple-differential cross sections

1s2s ^{3}S initial state, equal energy sharing

ω = 50 eV, $I = 5 \times 10^{14}$ Wcm^{-2}



- Two such contributions for $\theta_1 = 0^\circ$ and $\theta_1 = 90^\circ$.

Recoil-ion momentum distributions

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- Recoil-ion momentum vector given in terms of electron momenta:

$$\mathbf{p} = -\left(\mathbf{k}_1 + \mathbf{k}_2\right).$$

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- Recoil-ion momentum vector given in terms of electron momenta:

$$\mathbf{p} = -\left(\mathbf{k}_1 + \mathbf{k}_2\right).$$

- Momentum distribution obtained via integration of TDCS over appropriate set of angles and energies

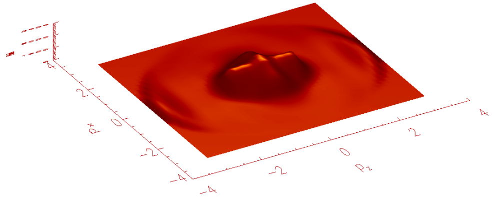
$$\frac{\mathrm{d}^3\sigma}{\mathrm{d}p_x \mathrm{d}p_y \mathrm{d}p_z} = \int \mathrm{d}\Omega_1' \int \mathrm{d}\Omega_2' \int \mathrm{d}E_1 \frac{\mathrm{d}^3\sigma}{\mathrm{d}\Omega_1 \mathrm{d}\Omega_2 \mathrm{d}E_1} \delta\left(\mathbf{p} + \mathbf{k}_1 + \mathbf{k}_2\right).$$

- Integration over all p_y gives distribution over p_x and p_z.

Recoil-ion momentum distributions

Singlet and triplet combined

$I = 5 \times 10^{14}$ Wcm^{-2}

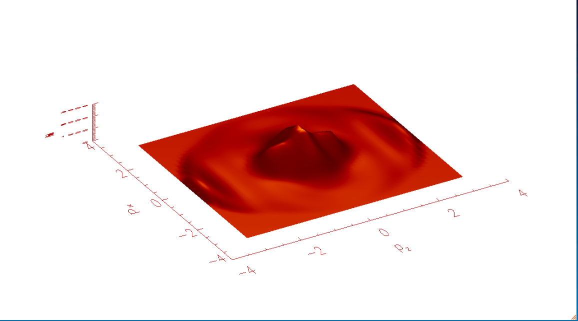


- Features at larger momenta $(|p_x, p_z| > 2)$ indicate three-photon double ionization.

Recoil-ion momentum distributions

Singlet and triplet combined

$I = 1 \times 10^{15}$ Wcm^{-2}



- Increase in magnitude of peak features associated with three-photon double ionization.

Recoil-ion momentum distributions

Singlet and triplet combined

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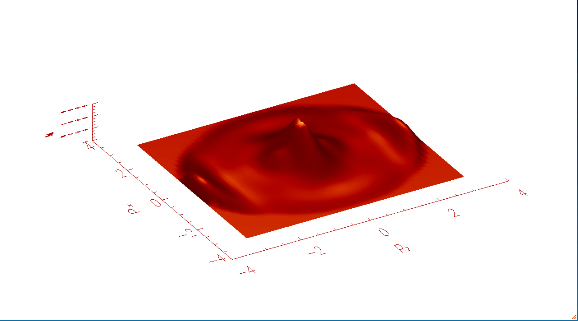

- Increase in central features $(p_z \simeq 0)$ associated with anti-parallel emission.

Comparison with experiment

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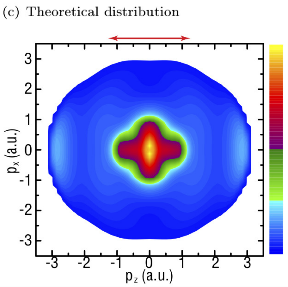
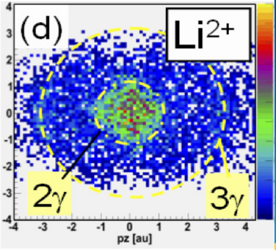
Comparison with experiment

TDCC
$I = 1 \times 10^{15}$ Wcm^{-2}



Experiment
(Schuricke/Dorn)



Singlet and triplet contributions

Two photon absorptions

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Singlet contribution

Two photon absorptions

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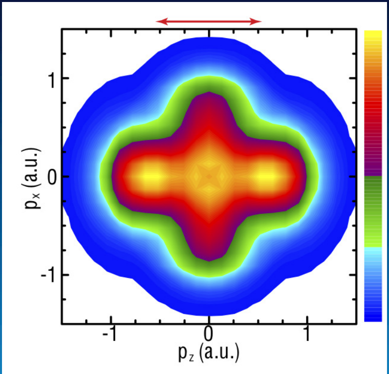
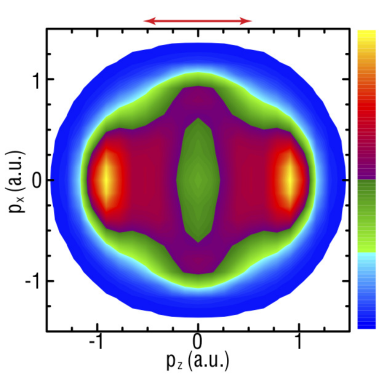


- Prominent central features due to anti-parallel emission.

Triplet contribution

Two photon absorptions

$I = 1 \times 10^{15}$ Wcm^{-2}



- Central minimum - anti-parallel emission largely avoided.

Double ionization of lithium at 59 eV

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- A minimum of two photons required for double ionization from an initial ground state.

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Double ionization of lithium at 59 eV

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- Double ionization proceeds via initial photoexcitation of the 1s2s2p $^2\mathrm{P}_{M=0}$ state.
- A single 59 eV photon is sufficient to ionize the 2s and 2p electrons.
- TDCC calculations model photoionization of 2s2p $^{1,3}\mathrm{P}_{M=0}$ two-electron states.

Triple-differential cross sections

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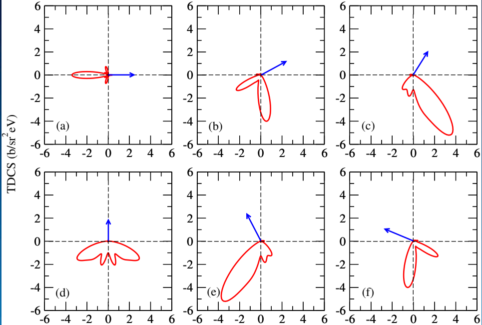
2s2p ^{1}P initial state, equal energy sharings

$\omega = 59$ eV, $I = 5 \times 10^{14}$ Wcm^{-2}



- Dominant anti-parallel emission for $\theta_1 = 0°$ with additional near-perpendicular components.

Triple-differential cross sections

2s2p ^{1}P initial state, equal energy sharings

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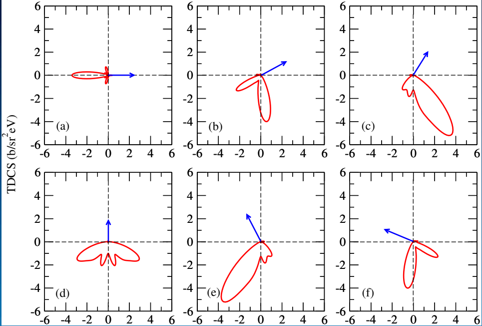
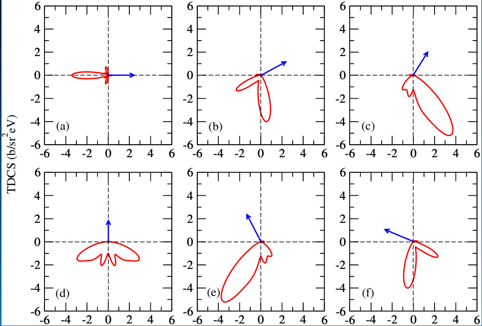


- Anti-parallel emission no longer dominant for $\theta_1 = 30°$ and $\theta_1 = 150°$.

Triple-differential cross sections

2s2p ^{1}P initial state, equal energy sharings

$\omega = 59$ eV, $I = 5 \times 10^{14}$ Wcm^{-2}



- Near-perpendicular emission dominant for $\theta_1 = 60^\circ$ and $\theta_1 = 120^\circ$.

Triple-differential cross sections

2s2p ^{1}P initial state, equal energy sharings

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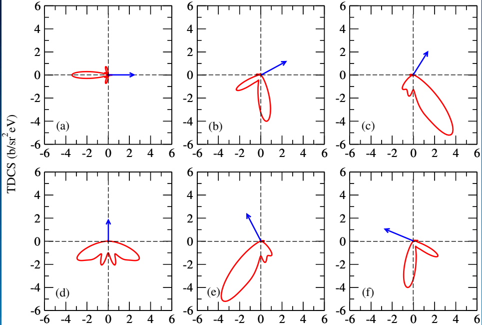
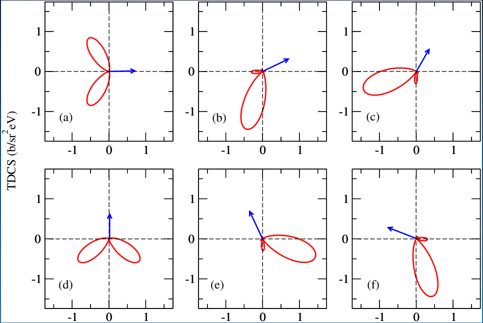


- Two peaks for $\theta_1 = 90^\circ$ at $\theta_2 = \theta_1 \pm 120^\circ$ with additional near-anti-parallel components.

Triple-differential cross sections

2s2p ^{3}P initial state, equal energy sharings

$\omega = 59$ eV, $I = 5 \times 10^{14}$ Wcm^{-2}



- Anti-parallel emission avoided for all values of θ_1.

Triple-differential cross sections

2s2p ^{3}P initial state, equal energy sharings

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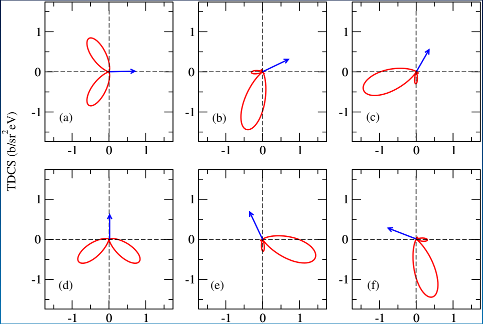

- Dominant emission at $\theta_{12} \simeq 130°$ at all values of θ_1.

Triple-differential cross sections

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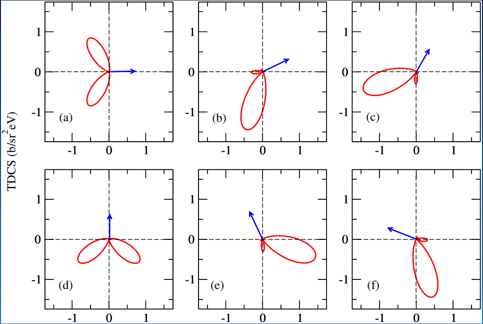

- Two such contributions for $\theta_1 = 0°$ and $\theta_1 = 90°$.

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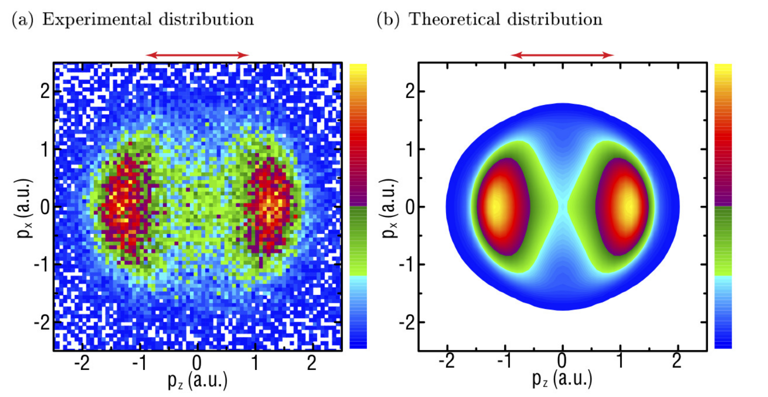
Recoil-ion momentum distributions

Comparison with experiment



- Reasonable agreement in lobe structure positions and relative magnitudes.

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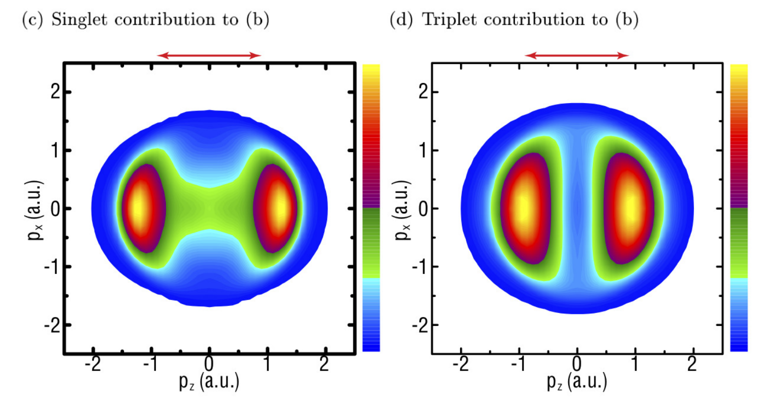
Singlet and triplet contributions



- Peaks at $p_z > 1$ in the singlet contribution indicate preference for emission into a common hemisphere.

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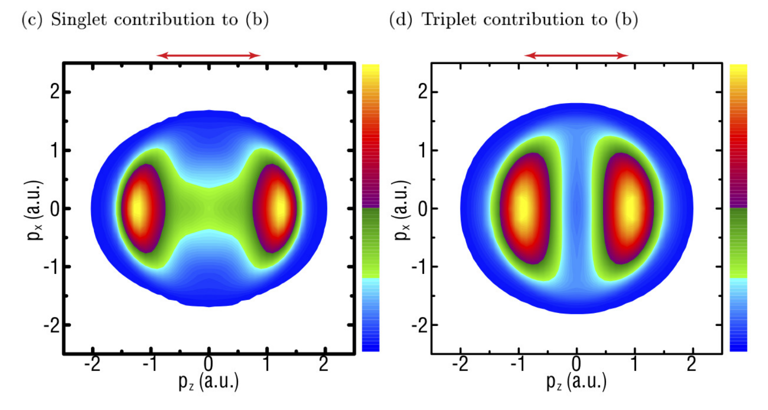
Singlet and triplet contributions



- Singlet contribution again shows central features due to anti-parallel emission.

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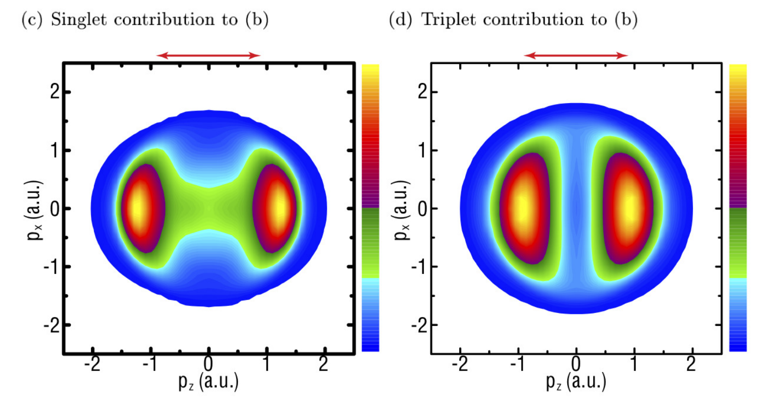
Singlet and triplet contributions



- No such features in the triplet contribution since anti-parallel emission is largely avoided.

Summary

- First set of calculations for two- and three-photon double ionization of Li.

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- Each spin state shows highly characteristic emission configurations.
- Intermediate photoexcitation has a considerable effect on emission configurations.
- Recoil-ion momentum distributions are in good agreement with FLASH measurements.

Electron-impact ionization of Na and Mg

Theory

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TDCC method

- Angular reduction of the two-electron time-dependent Schrödinger equation yields

$$i\frac{\partial}{\partial t}P^{LS}_{l_1l_2}(r_1,r_2,t) = T_{l_1l_2}P^{LS}_{l_1l_2}(r_1,r_2,t) + \sum_{l'_1,l'_2} V^{L}_{l_1l_2,l'_1l'_2}P^{LS}_{l'_1l'_2}(r_1,r_2,t).$$

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- Initial state is a spin-symmetrized product of an incoming electron wavepacket and a valence-shell radial orbital:

$$P^{LS}_{l_1 l_2}(r_1,r_2,t=0) = \sqrt{\frac{1}{2}}\left[G_{k_0 l_1}(r_1)P_{nl_2}(r_2) + (-1)^S P_{nl_1}(r_1)G_{k_0 l_2}(r_2)\right].$$

The challenge of multi-electron atoms

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- Accurate description of inactive multi-electron core and its interaction with active electrons required.

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- Higher angular momentum channels must be retained in the wavefunction for a more diffuse atomic system.

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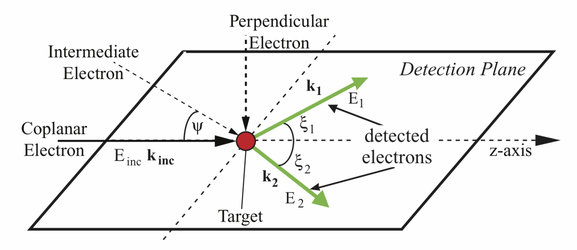
- Variable radial mesh required to track rapid oscillations of the radial wavefunction near the nucleus.

The challenge of multi-electron atoms

- Accurate description of inactive multi-electron core and its interaction with active electrons required.
- Higher angular momentum channels must be retained in the wavefunction for a more diffuse atomic system.
- Variable radial mesh required to track rapid oscillations of the radial wavefunction near the nucleus.
- Retain efficiency and accuracy using an appropriate time propagation scheme.

Triple-differential cross sections for electron-impact ionization of Na

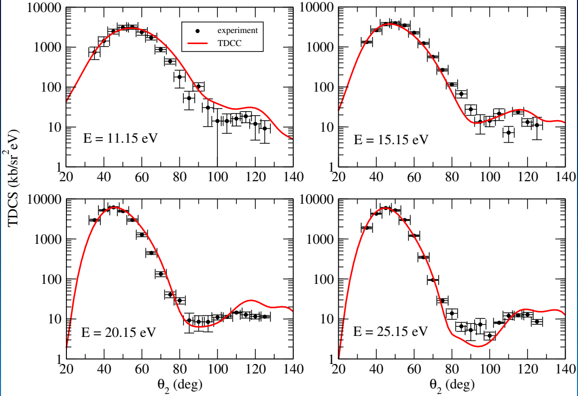
Manchester experiments



- Detection carried out in coplanar ($\phi_1 = \phi_2 = 0°$) symmetric ($\xi_1 = \xi_2$) or asymmetric geometries.

Triple-differential cross sections

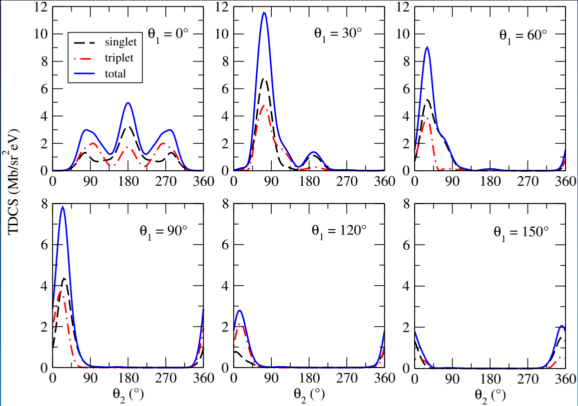
coplanar symmetric geometry, equal energy sharing



- Good agreement obtained with experiment in each case; calculations are able to track TDCS features spanning several orders of magnitude.

Triple-differential cross sections

coplanar asymmetric geometry, equal energy sharing, $E_{inc} = 11.15$ eV

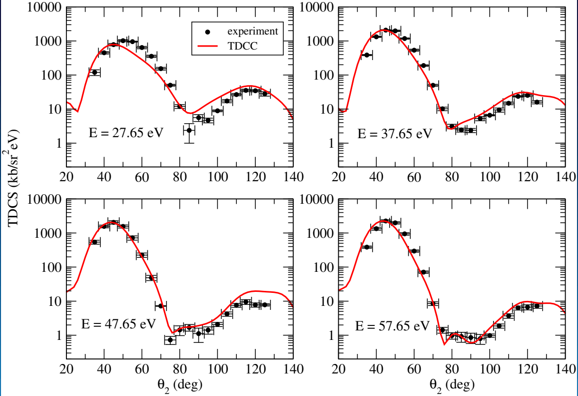


- Wannier breakup geometry dominates for $\theta_1 = 0°, 150°$.
- Overall preference for $\theta_1 = 30°$.
- Similar trends were observed in previous CCC calculations.

Triple-differential cross sections for electron-impact ionization of Mg

Triple-differential cross sections

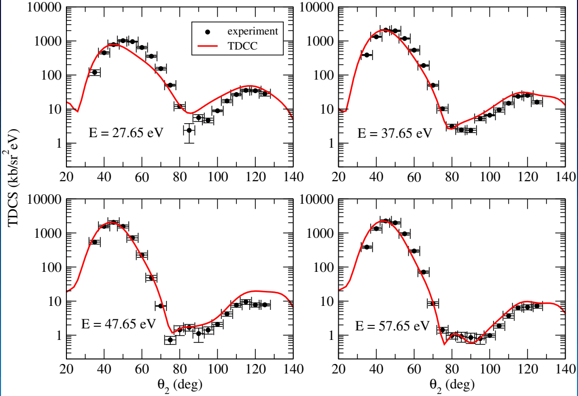
coplanar symmetric geometry, equal energy sharing



- Calculations are able to track TDCS features which range over several orders of magnitude.

Triple-differential cross sections

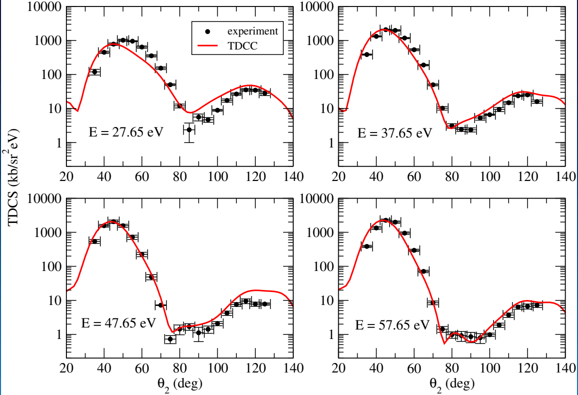
coplanar symmetric geometry, equal energy sharing



- A more complete treatment of valence-shell correlation may be required at 27.65 eV.

Triple-differential cross sections

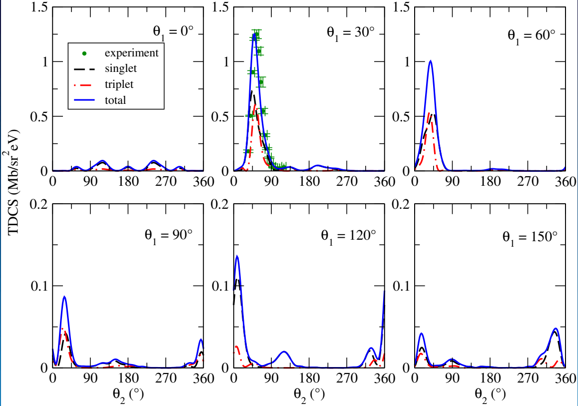
coplanar symmetric geometry, equal energy sharing



- Sensitivity to correlation may be reduced as the 3s electron is ejected more rapidly.

Triple-differential cross sections

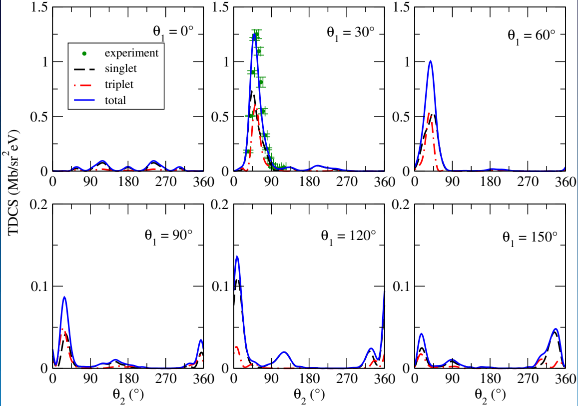
coplanar asymmetric geometry, equal energy sharing, $E_{inc} = 47.65$ eV



- Good agreement obtained with measurement at $\theta_1 = 30^\circ$.

Triple-differential cross sections

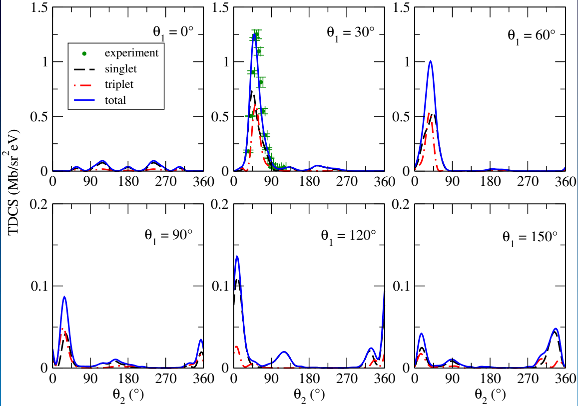
coplanar asymmetric geometry, equal energy sharing, $E_{inc} = 47.65$ eV



- Strong preference for emission with $\theta_1 = 30°$, as was the case for Na.

Triple-differential cross sections

coplanar asymmetric geometry, equal energy sharing, $E_{inc} = 47.65$ eV



- Additional structure in comparison to the Na TDCS.

Summary

- Applied a new version of the TDCC method to calculate angular distributions for ionization of multi-electron targets.
- Good agreement with measurements obtained for a range of incident electron energies for Na and Mg.
- Differing dynamics observed in the angular distributions for He and Mg at a common excess energy.

Future plans

- Calculate angular distributions for electron-impact single ionization of Mg where both valence-shell electrons are considered active.

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- Examine the effect of valence-shell correlation through comparison of two-electron and three-electron calculations.

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Future plans

- Calculate angular distributions for electron-impact single ionization of Mg where both valence-shell electrons are considered active.
- Examine the effect of valence-shell correlation through comparison of two-electron and three-electron calculations.
- Calculate angular distributions for electron-impact ionization of anistropic atomic targets and make comparison with recent experiments.

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